



A reduced mechanism for ethylene/methane mixtures with excessive NO enrichment

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ABSTRACT

A detailed mechanism for methane–ethylene mixtures enriched with excessive amount of NO was systematically reduced for efficient numerical simulations of flames in arc-heated co-flowing air. Methane and ethylene were selected as the surrogate fuel in the present study due to their drastically different features of ignition and extinction properties and flame propagation speeds, such that the mixtures of them may be utilized to mimic practical hydrocarbon fuels with various kinetic properties in experiments. The recently released USC Mech-II for C1–C4 was grafted with the NO_x sub-mechanism in GRI-Mech 3.0 with updated reaction parameters for prompt NO formation. The resulting detailed mechanism with 129 species and 900 reactions was first validated against experiments involving NO_x enrichment and reasonably good agreements were observed. The detailed mechanism was then employed as the starting mechanism for the reduction. A skeletal mechanism with 44 species and 269 reactions was derived using the methods of directed relation graph (DRG) and DRG-aided sensitivity analysis (DRGASA); a 39-species reduced mechanism with 35 semi-global reaction steps was further obtained using the linearized quasi steady state approximations (LQSSA). Five species related to prompt NO were retained in the reduced mechanism because of their significant impacts on the fuel oxidation. The reduced mechanism closely agrees with the detailed mechanism for ignition and extinction of homogenous mixtures, as well as selected 1-D flames over a wide range of parameters with NO concentrations between 0% and 3%. The observed worst-case relative error of the reduction is approximately 20%. The reduced mechanism was further validated with experiments involving excessive NO_x enrichment.

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1. Introduction

Lean blow-out (LBO) of non-premixed jet flames is an important topic on flame stabilization in scramjet research. Computational study of LBO is complicated by the complex chemical kinetics and the large number of components in practical jet fuels. In such cases, fuel components with dramatically different chemical properties, such as ignition delay, extinction time, and laminar flame speed, can be blended and serve as surrogates of practical jet fuels [1]. In particular, methane and ethylene feature drastically different chemical reactivities while sharing similar diffusivities. Moreover, the oxidation kinetics of methane and ethylene has been extensively studied and multiple detailed kinetic mechanisms have been developed, such as those in [2–4]. The blends of these two fuels may therefore be practically viable surrogates for efficient simulations of ignition, extinction, and flame propagation of jet fuel flames in scramjets.

Since arc-heaters were frequently utilized in scramjet experiments to preheat the co-flowing air, excessive NO may form through the thermal NO pathway in the high temperature zones near the arc-heater [5]. The effect of high NO concentration on fuel oxidation is nevertheless not totally clear. As such, numerical investigations with detailed chemistry for methane and ethylene mixtures with accurate NO reaction pathways can be an effective approach to understand the effect of NO on fuel oxidation. However, although detailed kinetic mechanisms are typically comprehensive and feature good accuracy, they are typically large and time-consuming for practical CFD simulations. For example, the detailed USC Mech-II for C1–C4 [2] consists of more than 100 species and approximately 800 reactions. Mechanism reduction is therefore necessary before such large mechanisms can be efficiently applied to CFD simulations of complex flow fields.

Mechanism reduction has been extensively studied and many methodologies have been developed in the last few decades [6]. Many of these methods fall into two major categories. The first is skeletal reduction, which eliminates unimportant species and reactions from the detailed mechanisms. The second is based on time

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scale analysis, which approximates exhausted fast processes with algebraic equations.

Skeletal reduction can be achieved through, for example, methods based on sensitivity analysis [7,8] and principle component analysis (PCA) [9]. Unimportant species can also be eliminated through Jacobian based methods [10–12]. These methods can be time-consuming for large-size mechanisms due to the large amount of rate evaluations in computing the sensitivity and Jacobian matrices. On the other hand, methods based on local reaction fluxes can be efficient to apply. Such methods include, for example, the detailed reduction [13], directed relation graph (DRG) [14], and DRG-based methods such as DRG with error propagation (DRGEP) [15,16], and direct path flux analysis method [17]. The DRG-based methods can be combined with sensitivity-based methods, e.g. DRG-aided sensitivity analysis (DRGASA) [18,19] and DRGEP and sensitivity analysis (DRGEP-SA) [20–22], to achieve an optimal balance between the reduction cost and the size of the resulting skeletal mechanism.

The second level of reduction involves time scale analysis, which approximates the differential description of fast reacting species with algebraic equations obtained by assuming that the fast chemical processes are exhausted. Such time scale analysis includes, for example, quasi steady state approximation (QSSA) [7,23–32], partial equilibrium approximation (PEA) [33,34], computational singular perturbation (CSP) [12,35–38], intrinsic low-dimensional manifold (ILDM) [39–41], pre-image curve (PIC) [42,43], and rate-controlled constrained equilibrium (RCCE) [44–48].

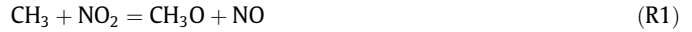
In the present study, a systematic approach outlined in [6], which integrated the DRG-based skeletal reduction and QSSA-based time scale reduction, is extended to reduce the detailed USC mechanism for methane/ethylene mixtures enriched with both low and high NO concentrations in a parameter range that is relevant to scramjet applications. The effect of NO enrichment on the reduced mechanism is further investigated.

2. Methodologies

2.1. The starting mechanism

In the present study, the detailed USC mechanism for C1–C4 oxidation [2] is employed as the starting mechanism for the reduction, grafted with the NO sub-mechanism from GRI 3.0 [49]. It is noted that while GRI3.0, including the NO_x subcomponent, is an optimized mechanism, the rate parameters were perturbed within the uncertainty ranges, such that the detailed chemical reaction paths were not substantially altered through the optimization [4]. As such it can be feasible to extract a subcomponent from a carefully optimized mechanism and merge it to different fuel oxidation mechanisms. Nevertheless, to account for the recent works on NO chemistry, rate parameters of five elementary reactions

involving NO formation were updated based on the literature data published after the release of GRI-Mech 3.0 [50–53]. A summary for the updated reaction rates is given in Table 1. The following reactions, R1 to R3, involving low temperature NO_x chemistry were further added into the detailed mechanism to account for the NO_x catalytic effect [54–56].



The resulting detailed mechanism consists of 129 species and 900 elementary reactions, which is substantially larger than the detailed mechanism for ethylene oxidation that was reduced in a previous work [14]. The detailed mechanism will be validated against experiment measurements later in Section 3.1.

The reduction of the detailed mechanism was performed within the parameter range of NO concentration from 0% to 3%, pressure from 0.5 to 10 atm, equivalence ratio from 0.5 to 2.0, initial temperature from 1000 to 1800 K for auto-ignition, and 700 K for extinction in perfectly stirred reactors (PSR). The fuel mixture consists of methane and ethylene in various ratios. Note that the parametric space for the reduction of binary fuel mixtures is one-dimension higher than that of single-component fuels, as such the sample space, and consequently the reduction cost, for fuel blends is substantially larger. Reaction conditions were sampled at approximately 100 discrete combinations of the above parameters, which densely cover the entire parameter range of interest, resulting in a large sample space consisting of about 6000 reaction states for auto-ignition and extinction in PSR. A local skeletal mechanism was obtained for each of the reaction states, and the union of all these local skeletal mechanisms constitutes the global skeletal mechanism that is valid over the entire parametric space. The worst-case error tolerance of the reduction was set to be 20%.

2.2. Skeletal reduction with DRG-based methods

The first step of the reduction is to eliminate unimportant species and reactions from the detailed mechanism and to obtain skeletal mechanisms. DRG was employed as the first reduction step for low computation cost and controllable reduction error [57]. In DRG, the coupling of a pair of species, *A* and *B*, was quantified as the relative error induced to *A* by the elimination of *B*:

$$r_{AB} \equiv \frac{\sum_{i=1,I} |v_{A,i} \omega_i \delta_{Bi}|}{\sum_{i=1,I} |v_{A,i} \omega_i|},$$

$$\delta_{Bi} = \begin{cases} 1, & \text{if the } i\text{th reaction involves } B \\ 0, & \text{otherwise} \end{cases}, \quad (1)$$

Table 1

List of the updated rate parameters for NO formation.

Reactions	Rate coefficients in GRI3.0			Updated rate coefficients		
	<i>A</i>	<i>n</i>	<i>E</i> (cal/mole)	<i>A</i>	<i>n</i>	<i>E</i> (cal/mole)
NCO + O = NO + CO	2.35E13	0	0	2.00E15 [50]	−0.50	0
NCO + OH = NO + H + CO	0.25E13	0	0	8.30E12 [51]	−0.10	75,488
HNCO + O = NCO + OH	2.20E06	2.11	11,400	2.20E06 [52]	2.11	47,823
HNCO + H = H ₂ + NCO	1.05E05	2.50	13,300	9.00E07 [52]	1.70	58,158
HCCO + NO = HCNO + CO	0.90E13	0	0	5.5E10 [53]	0	1830

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